

AMENDMENT UNDER 37 C.F.R. § 1.111  
U.S. Appln. No. 10/089,523

**REMARKS**

Applicants respectfully request the Examiner to confirm that the drawings filed with the application on March 29, 2002 have been accepted.

Also, Applicants respectfully request the Examiner to return initialed Form PTO-1449 from the Information Disclosure Statement filed June 18, 2003. Additionally, although citation to the Office Action issued October 7, 2002 in ROC Application No. 89120192 may be improper for listing on the front page of an issued patent, Applicants nonetheless submitted an English translation thereof, including citations to DE 199 09 574 and JP 683831, of record. Thus, all of the references cited in the ROC Office Action have been considered.

In accordance with the Examiner's request, Applicants submit herewith a substitute specification in proper idiomatic English complying with 37 C.F.R. § 1.52(a)(b). The substitute specification contains no new matter. The Abstract has also been amended for clarification.

Claims 1, 2 and 7 have been amended to recite --fluorine-containing resin fine particles having an average particle size of from 20 to 100 nm--. Support is found, for example, at page 10, line 5 of the specification.

Claims 8-10 and 14-16 have been canceled.

No new matter has been added, and thus entry of the Amendment is respectfully submitted to be proper. Upon entry of the Amendment, claims 1-7 and 11-13 will be all the claims pending in the application.

Applicants herein affirm the election of the invention of Group I, claims 1-5 and 11-13, for prosecution without traverse.

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Claims 1-7 and 11-13 were rejected under 35 U.S.C. § 102(b) as being anticipated by Logothetis et al (US 4,713,418).

Applicants respectfully submit that claims 1-7 and 11-13 as amended to require an average particle size of from 20 to 100 nm are not anticipated by Logothetis et al for the following reasons.

The thermoplastic particles of Logothetis et al have a particle size of "less than about 10 microns" (column 1, lines 64-66), and "[d]uring the cure and post-cure operations the thermoplastic tetrafluoroethylene copolymer phase separates into distinct particles, more or less spherical in shape, which are visible using an optical microscope" (column 2, lines 43-46).

Accordingly, the lower limit of the particle size of the thermoplastic resin particles of Logothetis et al is a particle size which is visible with an optical microscope.

The scientific theory of discrimination by an optical microscope is determined by the following equation:

$$d = (0.61 \times \lambda) / (n \times \sin \alpha)$$

where  $d$  is discrimination,  $\lambda$  is the wavelength of visible light,  $n$  is the refractive index of the lens,  $\alpha$  is angle of opening of incident light, and  $\sin \alpha$  is usually 1.5. Since the shortest wavelength of visual light is 0.4  $\mu\text{m}$ , according to the above equation, the maximum discrimination of an optical microscope is 0.2  $\mu\text{m}$  (200 nm). In order to observe finer structures, it is necessary to use an electron microscope.

As discussed above, the smallest particle size visible with an optical microscope is 200 nm. Therefore, because the thermoplastic resin fine particle of Logothetis et al is visible with an

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optical microscope, the particle size of the thermoplastic resin particle of Logothetis et al is more than 200 nm.

On the other hand, claims 1, 2 and 7 recite resin particles having an average particle size of from 20 to 100 nm. Accordingly, the present invention is not anticipated by Logothetis et al.

Further, Logothetis et al discloses that the tetrafluoroethylene copolymer is blended with an elastomer at a temperature sufficiently high to melt the tetrafluoroethylene copolymer (usually above 300 °C), and that the optically clear composition is obtained in a molten state in which the tetrafluoroethylene copolymer is at least partially dissolved in the elastomeric matrix (column 2, lines 31-37). After cooling, during the cure and post-cure operations, the thermoplastic tetrafluoroethylene copolymer phase separates into distinct particles (column 2, lines 38-45). Accordingly, Logothetis et al does not teach the present invention as recited in claim 1, even before curing.

Still further, as described above, the preparation process of Logothetis et al employs a molten blend. On the other hand, the process as recited in the present claim 6 is a co-coagulation blend process distinct from Logothetis et al.

In view of the above, the Examiner is respectfully requested to reconsider and withdraw the rejection.

Claims 1, 2, 4-7, 12 and 13 were rejected under 35 U.S.C. § 102(b) as being anticipated by Tung (US 4,348,312).

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Applicants respectfully submit that the present claims are not anticipated by Tung for the following reasons.

Tung discloses a transparent composition comprising a copolymer of methyl methacrylate (MMA) and ethyl methacrylate (EA), and an elastomeric copolymer of vinylidene fluoride (VdF) and hexafluoropropylene (HFP).

The MMA-EA copolymer is a thermoplastic resin but not a fluorine-containing resin. Further, Tung does not describe that the MMA-EA copolymer is in the form of particles. To the contrary, the present claims require fluorine-containing resin fine particles having an average particle size of from 20 to 100 nm.

According to Tung, the composition is prepared by mixing a xytol solution of MMA-EA copolymer and a xytol solution of VdF-HFP elastomer (Examples 77-85). Namely, Tung does not teach or suggest a co-coagulation process of mixing two emulsions as required by the present claim 6.

Thus, because Tung does not disclose one or more of the limitations of the rejected claims, it is respectfully submitted that the present invention is not anticipated by Tung, and thus the rejection should be withdrawn.

Claims 1-7 and 11-13 were rejected under 35 U.S.C. § 102(e) as being anticipated by Okanishi et al (US 6,476,144).

Applicants respectfully submit that claims 1-7 and 11-13 as amended are not anticipated by Okanishi et al for the following reasons.

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Okanishi et al discloses a "melt-processable crystalline fluorine-containing resin composition" (Abstract). The matrix polymer of Okanishi et al is a resin (column 3, lines 26-39), but not an elastomer. To the contrary, the present claims require an elastomer.

Thus, Okanishi et al fails to disclose one or more of the terms of the present claims, it is respectfully submitted that the present invention is not anticipated and thus the rejection should be withdrawn.

Withdrawal of rejections and allowance of claims 1-7 and 11-13 is earnestly solicited. If any points remain in issue which the Examiner feels may be best resolved through a personal or telephone interview, the Examiner is kindly requested to contact the undersigned at the telephone number listed below.

The USPTO is directed and authorized to charge all required fees, except for the Issue Fee and the Publication Fee, to Deposit Account No. 19-4880. Please also credit any overpayments to said Deposit Account.

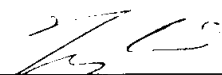
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## DESCRIPTION

## TRANSPARENT ELASTOMER COMPOSITION

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## TECHNICAL FIELD

The present invention relates to a transparent composition comprising an elastomer and fine particles of fluorine-containing resin, for example, a transparent elastomer composition comprising a fluorine-containing elastomer as a matrix and fine particles of fluorine-containing resin which are finely dispersed in the elastomer. The elastomer composition can provide a molded article ~~possessing~~ having enhanced mechanical strength and is useful as a base elastomer for various molded articles. Further, the elastomer composition can provide an elastomeric molded article which is excellent in mechanical strength and transparency by ~~selecting~~ adding an additive ~~to be added~~ thereto.

## BACKGROUND ART

It has been known that a fluorine-containing resin is added to a fluorine-containing elastomer. An object of the addition is to enhance ~~a~~-mechanical strength by using the fluorine-containing resin as a clean filler and ~~to utilize a~~ utilizing its friction property thereof. ~~Also Methods~~, for mixing the resin and the elastomer, ~~there are proposed include~~ a dry blend method by using a usual rubber mixing roll, and a method for enhancing ~~a~~-dispersibility by using a solvent, and the like.

For example, in JP-A-55-151051, a fluorine-containing rubber excellent in abrasion resistance is obtained by blending a low molecular weight polytetrafluoroethylene (PTFE). Also, JP-A-63-178149 describes a method of adding and mixing a PTFE powder to a fluorine-containing rubber solution ~~in order to~~ enhance ~~a~~ the gas barrier property and mechanical strength of ~~a~~ the rubber. Further, JP-A-2-261850 proposes that a fluorine-containing resin is blended in a

large amount of from 30 to 150 parts by weight together with an organic solvent to 100 parts by weight of a rubber to lower a friction coefficient and to enhance mechanical properties.

5        ~~An Example~~ example of an addition of a fluorine-containing resin ~~for to obtaining obtain~~ cleanliness is described in WO97/08239. WO97/08239 ~~which~~ discloses that a composition prepared by adding 5 to 50 parts by weight of a fluorine-containing resin powder having an average particle size of from 0.2 to 50  $\mu$ m to 100 parts by weight of a fluorine-containing rubber is excellent as a sealing agent for etching  
10       equipment. Also, WO95/02634 discloses that a composition prepared by adding 2 to 50 parts by weight of a fluorine-containing resin fine powder to 100 parts by weight of a rubber component is a clean composition suitable for a wet process of semiconductors.

15       However, those prior techniques are directed to ~~addition~~ adding and mixing ~~of~~ a fluorine-containing resin to a matrix rubber (elastomer). ~~and even if a~~ Although the dispersibility ~~is~~ may be enhanced by using a solvent, as a particle size becomes finer, a uniform dispersion is difficult to obtain. ~~This is shown by a fact that~~  
20       ~~for~~ For example, when fine particles of a fluorine-containing resin are dispersed in a transparent elastomer, if they are dispersed uniformly, the composition should be transparent. Yet, ~~but actually~~ a transparent elastomer composition in which a fluorine-containing resin is finely uniformly dispersed has not yet been obtained.

25       If fine particles of a fluorine-containing resin are not dispersed uniformly, a strength of a molded article obtained by vulcanizing and molding is low and ~~an~~ the effect of adding the fluorine-containing resin is not obtained sufficiently.

30       It is therefore an object of the present invention ~~is to~~ provide an elastomer composition ~~which is in a state that~~ wherein fluorine-containing resin fine particles are dispersed uniformly in a transparent elastomer, and which can exhibit the maximum ~~of~~ effect of adding the fluorine-containing resin.

## DISCLOSURE OF INVENTION

Namely, ~~the~~ The present invention relates to a transparent elastomer composition in which fine particles of a fluorine-containing resin are finely dispersed in a transparent elastomer.

It is preferable that the transparent elastomer as a matrix is a fluorine-containing elastomer. Also, it is preferable that the fine particles of a fluorine-containing resin have an average particle size of from 20 to 150 nm.

The present invention also relates to an elastomer composition containing a crosslinking agent and as ~~the ease demands required~~, a crosslinking accelerator and a filler.

The elastomer composition of the present invention can be prepared, for example, by mixing an emulsion of transparent elastomer particles and an emulsion of fluorine-containing resin fine particles and then co-coagulating the resulting mixture.

The present invention also relates to an elastomeric molded article obtained by vulcanizing and molding the above-mentioned elastomer composition, ~~and~~. The present invention further relates to a transparent elastomeric molded article.

In the present invention, being transparent means that a haze value mentioned hereinbelow is not more than 50 %.

## BRIEF DESCRIPTION OF DRAWINGS

Fig. 1 is a photograph ( $\times 20000$ ) taken by a transmission type electron microscope of the elastomer composition obtained in Example 1 of the present invention.

Fig. 2 is a photograph ( $\times 20000$ ) taken by a transmission type electron microscope of the elastomer composition obtained in Comparative Example 3.



## BEST MODE FOR CARRYING OUT THE INVENTION

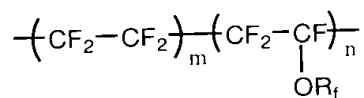
In the present invention, uniform dispersion of fine particles of a fluorine-containing resin ~~which has been difficult to obtain~~ is achieved by ~~the method of mixing~~ an emulsion of transparent elastomer particles and an emulsion of fluorine-containing resin fine particles and then co-coagulating the mixture.

Whether or not the fluorine-containing resin fine particles are dispersed finely uniformly can be seen ~~obviously~~ clearly by observing through a transmission type electron microscope (TEM) (cf. Fig. 1 explained hereinbelow) or can be judged by determining whether or not transparency of the transparent matrix elastomer is maintained even after mixing of the fluorine-containing resin fine particles. In other words, the mixture obtained by dispersing is turbid in white, even if the dispersing is carried out, either by a dry blend method in which the mixing is carried out after the fluorine-containing resin fine particles having the same fine particle size are once coagulated and dried, or by a method of mixing by using a solvent. It ~~can be considered~~ is believed that this is because the fluorine-containing resin fine particles are subject to secondary agglomeration or are dispersed locally (cf. Fig. 2 explained hereinbelow).

The elastomer composition of the present invention before crosslinking has a transparency of not more than 50 %, preferably not more than 40 %, particularly not more than 30 % in a haze value.

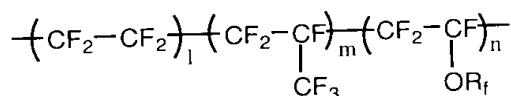
~~The~~ For the matrix elastomer ~~to be used~~ in the present invention, ~~is good as far as it is a~~ any transparent elastomer capable of forming an emulsion in the form of particles and ~~has~~ having affinity for the fluorine-containing resin can be used. From this point of view, a fluorine-containing elastomer is preferable.

Examples of the fluorine-containing elastomer are, for instance, copolymer elastomers (tetrafluoroethylene (TFE)/perfluoro(alkyl vinyl ether) (PAVE) elastomers) represented by the formula (1):

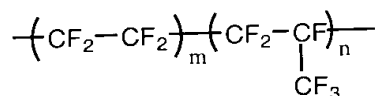


wherein  $m/n = 95$  to  $50/5$  to  $50$  (% by mole, hereinafter the same),  $R_f$  is a perfluoroalkyl group having 1 to 8 carbon atoms,

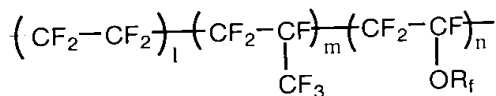
- 5 terpolymer elastomers (TFE/hexafluoropropylene (HFP)/PAVE elastomers) represented by the formula (2):



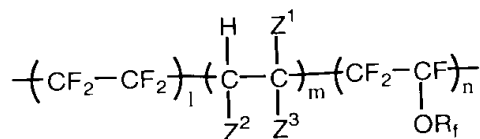
- 10 wherein  $1/m/n = 95$  to  $35/0$  to  $30/5$  to  $35$ ,  $R_f$  is a perfluoroalkyl group having 1 to 8 carbon atoms, copolymer elastomers represented by the formula (3):



- 15 wherein  $m/n = 85$  to  $60/15$  to  $40$ , terpolymer elastomers represented by the formula (4):

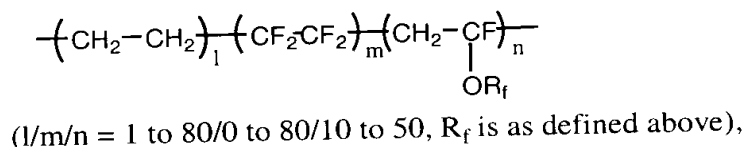
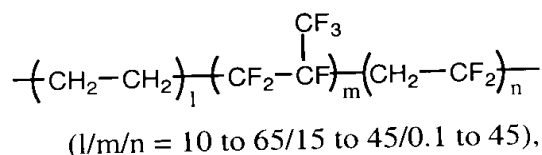
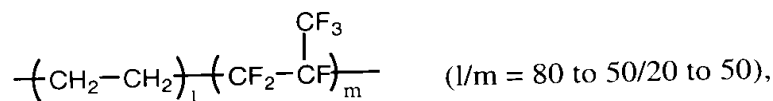
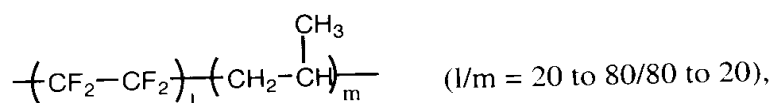


wherein  $1/m/n = 85$  to  $20/0$  to  $40/15$  to  $40$ , terpolymer elastomers represented by the formula (5):



- 20 wherein  $1/m/n = 95$  to  $45/0$  to  $10/5$  to  $45$ ,  $Z^1$ ,  $Z^2$  and  $Z^3$  are fluorine

atoms or hydrogen atoms independently, R<sub>f</sub> is a perfluoroalkyl group having 1 to 8 carbon atoms.



5

and the like.

More ~~concretely~~ specifically, there are TFE/PAVE copolymer elastomer, vinylidene fluoride (VdF)/hexafluoropropylene (HFP) copolymer elastomer, VdF/HFP/TFE copolymer elastomer, VdF/TFE/PAVE copolymer elastomer, and the like. A small amount of a monomer having a crosslinkable reaction group may be copolymerized with those elastomers. Examples of the crosslinkable reaction group are, for instance, iodine atom, bromine atom, nitrile group, carboxyl group, unsaturated double bond, hydroxyl group, and the like.

~~These~~ The fluorine-containing elastomers can be prepared by usual emulsion polymerization method, ~~and the~~ The obtained polymerization reaction product which is an emulsion can be used as it is for co-coagulation to be described hereinbelow, or can be used for the co-coagulation after optionally adjusting ~~a~~ the concentration thereof. The emulsion may be ~~one~~ dried and then formed into an

20

dispersion again.

Examples of the elastomer other than the fluorine-containing elastomer are, ~~for instance,~~ hydrogenated nitrile butadiene rubber, acrylic rubber, silicone rubber, and the like.

5           An average particle size of the elastomer particles in the emulsion is not ~~limited~~-particularly limited, and is, for example, from 10 to 800 nm, preferably from 20 to 500 nm. However, when the average particle size is less than 10 nm, the coagulation becomes difficult, and when the average particle size is more than 800 nm, the  
10       emulsion becomes unstable and the co-coagulation becomes difficult.

The fine particles of the fluorine-containing resin to be finely dispersed into the elastomer are not ~~limited~~-particularly limited. For example, there are fine particles of:

(1) polytetrafluoroethylene (PTFE);

15       (2) TFE/ $\text{CF}_2=\text{CF}-\text{O}-\text{R}_f^1$  (FVE) copolymer (~~Proportion thereof is one exhibiting a non-elastomeric property, for example, a proportion of in a proportion exhibiting a non elastomeric property for example, wherein the non-elastomer component, i.e.,~~  $\text{CF}_2=\text{CF}-\text{O}-\text{R}_f^1$  is not more than 15 % by mole.  $\text{R}_f^1$  is a linear or branched fluoro- or perfluoroalkyl group  
20       or a fluoro- or perfluorooxyalkyl group which may have an ether type oxygen atom.). For example, TFE/PAVE copolymer (PFA);

(3) TFE/ $\text{CF}_2=\text{CF}-\text{R}_f^1$  copolymer (~~Proportion thereof is one exhibiting a non-elastomeric property, for example, a proportion of in a proportion exhibiting a non elastomeric property for example, wherein the non-~~  
25       elastomer component, i.e.,  $\text{CF}_2=\text{CF}-\text{R}_f^1$  is not more than 15 % by mole.  $\text{R}_f^1$  is as defined above.) For example, TFE/HFP copolymer (FEP);

(4) Ethylene/TFE (30 to 60/70 to 40% by mole-in-% by mole, ~~hereinafter the same~~) copolymer;

(5) Polychlorotrifluoroethylene (PCTFE);

30       (6) Ethylene/chlorotrifluoroethylene (CTFE) (30 to 60/70 to 40% by mole) copolymer;

- (7) Polyvinylidene fluoride (PVdF);  
(8) Vinylidene fluoride (VdF)/TFE (70 to 99/30 to 1% by mole) copolymer;  
(9) VdF/TFE/CTFE (50 to 99/30 to 0/20 to 1% by mole) copolymer;  
5 (10) VdF/TFE/HFP (60 to 99/30 to 0/10 to 1% by mole) copolymer;  
(11) Ethylene/TFE/HFP (6 to 60/40 to 81/1 to 30% by mole) copolymer;  
(12) 3,3,3-trifluoropropylene-1,2-trifluoromethyl-3,3,3-trifluoropropylene-1/PAVE (40 to 60/60 to 40% by mole) copolymer;  
10 and the like. Among them, when a low friction property is imparted to the molded article, the above-mentioned (1) is preferred. Particularly, in order to enhance compatibility with the perfluoroelastomer component, the above-mentioned (2) and (3) are preferred.

The PTFE of the above-mentioned (1) encompasses not only  
15 TFE homopolymer but also a modified PTFE obtained by copolymerizing a comonomer in such a small amount as not giving melt-flowability. Examples of the comonomer are HFP, CTFE, perfluorovinylether, trifluoroethylene, perfluoroalkylethylene, and the like. When perfluorovinylether is ~~copolymerized~~ used as a comonomer,  
20 its amount is up to 2 % by weight, preferably from 0.001 to 1 % by weight, more preferably from 0.01 to 1 % by weight.

~~These~~ The fluorine-containing resins can be prepared by usual emulsion polymerization method, ~~and the~~ The obtained polymerization ~~reaction~~ product which is an emulsion can be used as it  
25 is for co-coagulation to be described hereinbelow, or can be used for the co-coagulation after optionally adjusting a the concentration thereof. The emulsion may be once dried and then formed into an emulsified dispersion again.

An average particle size of the fluorine-containing resin fine  
30 particles in the emulsion is less than 150 nm, preferably from 20 to 150 nm, more preferably from 20 to 100 nm. When the average particle size is too small, productivity is lowered remarkably, and when

the average particle size is not less than 200 nm, a uniform dispersion cannot be obtained.

The mixing ratio of the elastomer and the fluorine-containing resin may be selected optionally depending on properties to be imparted to the molded article. It is desirable that the amount of the fluorine-containing resin is not less than 1 part by weight, preferably not less than 5 parts by weight based on 100 parts by weight of the elastomer from the viewpoint of obtaining a reinforcing effect, and is not more than 150 parts by weight, preferably not more than 100 parts by weight, more preferably not more than 50 parts by weight based on 100 parts by weight of the elastomer from the viewpoint of easiness in processing of the obtained crosslinked rubber.

The ~~combination of the elastomer with and~~ the fluorine-containing resin in the combination may be selected ~~to intended functions in with the~~ consideration of that whether or not coagulating property thereof is approximate to each other and whether or not those polymers have affinity.

~~Non-restricted examples~~ Non-limiting examples of the preferable combination ~~are, for instance, include:~~

- (a) TFE/PAVE elastomer with TFE/PAVE resin (PFA);
  - (b) TFE/PAVE elastomer with TFE/HFP resin (FEP);
  - (c) VdF/HFP elastomer with PVdF resin;
  - (d) VdF/HFP/TFE elastomer with PVdF resin;
  - (e) TFE/propylene elastomer with ethylene/TFE resin (ETFE);
- and the like.

The elastomer composition of the present invention can be prepared by mixing the above-mentioned emulsion of the elastomer particles to the emulsion of the fluorine-containing resin fine particles and then coagulating the mixture. The co-coagulation can be carried out by a known coagulation method. For example, ~~there can be used a~~ method of adding a solution of the mixture of the emulsion of the elastomer particles and the emulsion of the fluorine containing resin

fine particles dropwise into a coagulating solution, or a method of adding a coagulating solution dropwise into a solution of the emulsion mixture, or the like method can be used.

5 The concentration of the emulsion mixture may be determined optionally depending on productivity, and is not particularly limited~~particularly~~. The concentration is usually from 5 to 50 % by weight, preferably from 10 to 30 % by weight. It is possible that the emulsion mixture is diluted with pure water 2 to 10 times before the coagulation.

10 ~~Concretely there are~~ Examples of the co-coagulation methods, ~~for instance, include~~ a salting out method, an acid coagulation method, a freeze coagulation method, a method of applying a mechanical shearing force, and the like.

15 As a coagulating agent, there can be used, for example, an acid such as ~~a~~-nitric acid, hydrochloric acid or sulfuric acid; a metal salt such as aluminum nitrate or aluminum sulfate; or the like. Among them, the acid is preferred from the viewpoint of keeping the polymer clean, and the metal salt is preferred from the viewpoint of easiness of handling.

20 A product obtained by the co-coagulation is washed as ~~the ease demands~~required, and dried in a hot air dryer or a vacuum dryer, and thus can be used as a base material for molding.

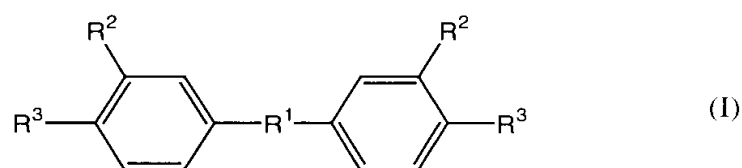
25 When the elastomer composition of the present invention is used as a base material, excellent mechanical strength, abrasion resistance, transparency and mold-processability can be imparted to the obtained molded article.

To the elastomer composition of the present invention can be added a crosslinking agent and further a crosslinking accelerator, and thus a crosslinkable elastomer composition can be obtained.

30 As a crosslinking system, those which have been usually used for an elastomer can be used. Examples thereof are, ~~for instance,~~ an oxazole crosslinking system, imidazole crosslinking system, thiazole

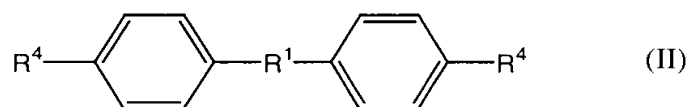
crosslinking system, triazine crosslinking system, peroxide crosslinking system, polyol crosslinking system, polyamine crosslinking system, and the like. Also, crosslinking with radiant ray, electron beam, ultraviolet ray, or the like can be carried out.

- 5 Examples of the crosslinking agent which is used in oxazole crosslinking system, imidazole crosslinking system and thiazole crosslinking system are, ~~for instance,~~ a tetraamine crosslinking agent or bisamino(thio)phenol crosslinking agent represented by the formula

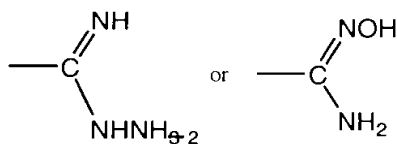


- 10 wherein R<sup>1</sup> is -SO<sub>2</sub>-, -O-, -CO-, an alkylene group having 1 to 6 carbon atoms, a perfluoroalkylene group having 1 to 10 carbon atoms or a single bond, one of R<sup>2</sup> and R<sup>3</sup> is -NH<sub>2</sub> and ~~another~~ the other one is -NH<sub>2</sub>, -OH or -SH, a bisamidrazone crosslinking agent represented by the formula (II):

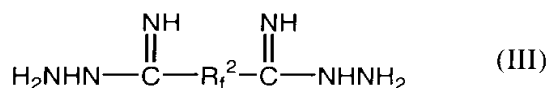
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(wherein R<sup>1</sup> is as defined above, R<sup>4</sup> is

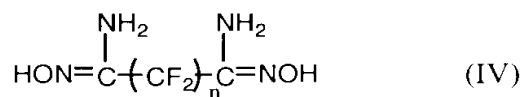


- 20 and a bisamidoxime crosslinking agent represented by the formula (III) or (IV):



wherein R<sub>f</sub><sup>2</sup> is a perfluoroalkylene group having 1 to 10 carbon atoms,





wherein n is an integer of 1 to 10, and the like.

As the case demands, a—crosslinking accelerator accelerators may be used together in combination.

5 An amount of the crosslinking agent is from 0.5 to 10 parts by weight, preferably from 1 to 5 parts by weight based on 100 parts by weight of the elastomer, and an amount of the crosslinking accelerator is from 0.1 to 10 parts by weight, preferably from 0.2 to 5 parts by weight based on 100 parts by weight of the elastomer.

10 Examples of the crosslinking agent to be used in the peroxide crosslinking system are, for instance, include 1,1-bis(t-butylperoxy)-3,5,5-trimethylcyclohexane trimethylcyclohexane, 2,5-dimethylhexane dimethylhexane -2,5-dihydroperoxide, di-t-butylperoxide, t-butylcumylperoxide, dicumylperoxide, α,α-bis(t-butylperoxy)-p-diisopropylbenzene, 2,5-dimethyl-2,5-di(t-butylperoxy)hexane, 2,5-dimethyl-2,5-di(t-butylperoxy)-hexyne-3, benzoylperoxide, t-butylperoxybenzene, 2,5-dimethyl-2,5-di(benzoylperoxy)hexane, t-butylperoxymaleate, t-butylperoxyisopropyl carbonate, and the like.

20 In case of the peroxide crosslinking, it is desirable to use a crosslinking accelerator. Examples of the crosslinking accelerator are, for instance, triallyl cyanurate, triallyl isocyanurate, triallyl formal, triallyl trimellitate, N,N'-m-phenylenebismaleimide, dipropargyl terephthalate, diallyl phthalate, tetraallyl terephthalateamide, triallyl phosphate, and the like.

25 An amount of the crosslinking agent is from 0.05 to 10 parts by weight, preferably from 1.0 to 5 parts by weight based on 100 parts by weight of the elastomer, and an amount of the crosslinking accelerator is from 0.1 to 10 parts by weight, preferably from 0.5 to 5

parts by weight based on 100 parts by weight of the elastomer.

A transparent elastomeric molded article can be provided in case where, like the peroxide crosslinking system, an inorganic filler, etc. does not need to be used for crosslinking, coloring does not occur  
5 through crosslinking and a vulcanized article from a so-called pure rubber formulation exhibits a transparency.

The transparent elastomeric molded article of the present invention obtained by crosslinking and molding has a transparency of not more than 50 %, preferably not more than 40 %, particularly not  
10 more than 30 % in a haze value.

Examples of the crosslinking agent which can usually be used in the polyol crosslinking system are aromatic polyhydroxy compounds ~~to be usually used~~. For example, there are 2,2-bis(4-hydroxyphenyl)propane (so-called "bisphenol A"), 2,2-bis(4-hydroxyphenyl)perfluoropropane (so-called "bisphenol AF"), resorcin,  
15 1,3,5-trihydroxybenzene, 1,7-dihydroxynaphthalene, 2,7-dihydroxynaphthalene, 1,6-dihydroxynaphthalene, 4,4'-dihydroxydiphenyl, 4,4'-dihydroxystilbene, 2,6-dihydroxyanthracene, hydroquinone, catechol, 2,2-bis(4-hydroxyphenyl)butane (so-called  
20 "bisphenol B"), 4,4-bis(4-hydroxyphenyl)valerate, 2,2-bis(4-hydroxyphenyl)tetrafluorochloropropane, 4,4'-dihydroxydiphenylsulfone, 4,4'-dihydroxydiphenylketone, tri(4-hydroxyphenyl)methane, 3,3',5,5'-tetrachlorobisphenol A, 3,3',5,5'-tetrabromobisphenol A, alkali metal salt or alkali earth metal salt  
25 thereof, and the like.

Also, in the polyol crosslinking system, it is preferable to ~~use-add~~ a crosslinking accelerator ~~together~~. Examples thereof ~~are, for instance,~~ include ammonium compounds, phosphonium compounds, oxonium compounds, sulfonium compounds, and the like.  
30 Particularly, tertiary ammonium salt and tertiary phosphonium salt are preferable.

An amount of the crosslinking agent is from 0.5 to 5 parts

by weight based on 100 parts by weight of the elastomer, and an amount of the crosslinking accelerator is from 5 to 400 parts by weight, preferably from 10 to 100 parts by weight based on 100 parts by weight of the crosslinking agent.

5           As a crosslinking agent to be used for the polyamine crosslinking, there are polyamine compounds. ~~Examples of the~~ The polyamine compound ~~are~~ is a primary amine or secondary amine in which two or more basic nitrogen atoms are bonded in ~~a~~ the molecule thereof. In many cases, ~~those amines are~~ the amine is modified to be  
10 ~~formed into~~ a salt so that a reaction becomes mild. Examples thereof are, ~~for instance,~~ include alkylenediamines such as ethylenediamine carbamate, hexamethylenediamine carbamate and 4,4'-diaminocyclohexylmethane carbamate which are used comparatively well. Also, ~~there can be used a~~ Schiff base, can be used such as N,N'-  
15 dicinnamylidene-1,6-hexamethylenediamine. In addition, the aromatic polyamine compounds having a poor basic property can be used preferably in combination with other basic compounds. Examples of the other basic compound are, ~~for instance,~~ include  
20 diphenylguanidine, di-o-triguanidine, diphenylthiourea, 2-mercaptoimidazoline, ~~and in addition,~~ a compound which has -NH<sub>2</sub> and/or -NH- in its molecule and is used as a crosslinking accelerator for a synthetic rubber, a divalent metal hydroxide, and the like.

          An ~~adding~~ addition amount of the crosslinking agent is from 0.1 to 10 parts by weight, preferably from 0.5 to 5 parts by weight  
25 based on 100 parts by weight of the elastomer.

          Examples of the other additives are a filler (excluding the above-mentioned fluorine-containing resin), a pigment, and the like.

          Examples of the filler ~~are, for instance,~~ include inorganic fillers such as carbon black (particularly graphitized carbon black),  
30 silicon oxide, titanium oxide and alumina; organic fillers such as polyimide, and the like. An adding amount thereof is not more than

100 parts by weight, preferably from 1 to 50 parts by weight based on 100 parts by weight of the elastomer.

5 The above-mentioned additives such as a crosslinking agent, a crosslinking accelerator and a filler may be added at the time of the above-mentioned co-coagulation, if possible, or may be mixed after the preparation of the composition comprising the elastomer and the fluorine-containing resin fine particles. The mixing method may be a known kneading method by using a roll, or the like ~~method~~.

10 A crosslinked molded article can be produced by kneading and crosslinking the thus obtained crosslinkable elastomer composition in which fluorine-containing resin fine particles are finely dispersed. For the kneading, usual methods, for example, kneading by a roll or kneader can be employed. For the molding, usual compression molding, injection molding, extrusion molding and transfer molding  
15 methods can be employed. The molding conditions may be the same as conventional ones.

The obtained crosslinked molded article maintains transparency of the matrix elastomer when a filler is not contained therein. Though there is a case that the transparency is lowered  
20 somewhat, a transmissivity of visible light is maintained at 20 % or more of that ~~in case of sole use of the elastomer alone~~.

The crosslinked molded article of the present invention is excellent in mechanical strength, post-processability, plasma resistance and gas barrier property, irrespective of presence or absence  
25 of ~~the~~ a filler. Also, since the finely dispersed fluorine-containing resin fine particles ~~finely dispersed~~ are hardly separated from the matrix elastomer, for example, even if the molded article is used as a sealing material for semiconductor production apparatuses, there is less possibility of generating micro-particles.

30 The elastomer composition of the present invention is ~~suitably used~~ suitable for various molded articles shown in Tables 1 to 3 ~~by making the use of~~ due to its excellent properties.

TABLE 1

Field of industry	Sub-field of industry	Final product	Equipment
Electrical	Semi-conductor	Semi-conductor production apparatus	CVD equipment
		Liquid crystal panel production apparatus	Dry etching equipment
		Plasma panel production apparatus	Wet etching equipment
			Oxidation and diffusion equipment
			Sputtering equipment
			Ashing equipment
Transportation	Vehicle		Cleaning equipment
			Ion implantation equipment
		Car	Engine and peripheral equipment
			AT
			Fuel line and peripheral equipment
		Aircraft	Fuel line
		Rocket	Fuel line
		Ship	Fuel line

- continued -

- continued -

Parts

O (square) ring, packing, sealing material, tube, roll, coating, lining, gasket, diaphragm, hose

Gasket, shaft seal, valve stem seal, sealing material, hose

Hose, sealing material

O (square) ring, tube, packing, core material of valve, hose, sealing material, diaphragm

Diaphragm, O (square) ring, valve, tube, packing, hose, sealing material

ditto

ditto

- continued -

- continued -

Chemical	Chemical products	Plant	Production line of chemicals such as pharmaceutical, agricultural chemical, coating and resin
			(Petroleum)
	Chemicals	Pharmaceuticals	Plug for chemicals
Mechanical	Photograph	Developing machine	Film developing machine
			X-ray film developing machine
	Printing	Printing machine	Printing roll
	Coating	Coating facilities	Coating roll
	Analyzer and physical and chemical appliances		
Food	Plant		Food processing line
Metal	Steel making	Steel sheet processing facilities	Steel sheet processing roll

- continued -

- continued -

Lining, valve, packing, roll, hose, diaphragm, O (square) ring, tube, sealing material

Plug for chemicals

Roll

Roll

Roll

Roll

Tube

Lining, valve, packing, roll, hose, diaphragm, O (square) ring, tube, sealing material

Roll



TABLE 2

Field of Industry	Characteristics Required
Electrical	Plasma resistance, acid resistance, alkali resistance, amine resistance, ozone resistance, gas resistance, chemical resistance, cleanliness, heat resistance
Transportation	Heat resistance, amine resistance
	Heat resistance, amine resistance
	Fuel resistance, fuel permeability, heat resistance
	Fuel resistance, fuel permeability, heat resistance
	Fuel resistance, fuel permeability, heat resistance
	Fuel resistance, fuel permeability, heat resistance
	- continued -

- continued -

Chemical	Chemical resistance, solvent resistance, heat resistance
	Chemical resistance, solvent resistance, heat resistance
	Cleanliness
Mechanical	Chemical resistance
	Chemical resistance
	Solvent resistance
	Solvent resistance
Food	Chemical resistance, solvent resistance, heat resistance
Metal	Heat resistance, acid resistance

TABLE 3

Field of industry	Parts
Electrical	O ring and sealing material for gate valve of corresponding production equipment
	O ring and sealing material for quartz window of corresponding production equipment
	O ring and sealing material for chamber of corresponding production equipment
	O ring and sealing material for gate of corresponding production equipment
	O ring and sealing material for bell jar of corresponding production equipment
	O ring and sealing material for coupling of corresponding production equipment
	O ring and sealing material for pump of corresponding production equipment
	O ring and sealing material for gas controller for semi-conductor of corresponding production equipment
	O ring and sealing material for resist developing and releasing solutions
	O ring and sealing material for wafer cleaning solution
	Diaphragm of pump for corresponding production equipment
	Hose for resist developing and releasing solutions
	Hose and tube for wafer cleaning solution
	Roll for transferring wafer
	Lining and coating of tanks for resist developing and releasing solutions
	Lining and coating of tanks for wafer cleaning solution
	Lining and coating of tanks for wet etching

- continued -

Engine head gasket

## Crank shaft seal

## Valve stem seal

Oil hose

## Injector O ring

### O ring and diaphragm for fuel pump

- continued -

- continued -	
Chemical	
Mechanical	Developing roll
	Developing roll
	Gravure roll
	Guide roll
	Gravure roll for magnetic tape production and coating line
	Guide roll for magnetic tape production and coating line
	Various coating rolls
Food	
Metal	

Particularly the molded articles of the present invention can be ~~used~~—built-in the following semiconductor manufacturing equipment.

(1) Etching system

5        Dry etching equipment

         Plasma etching machine

         Reactive ion etching machine

         Reactive ion beam etching machine

         Sputter etching machine

10       Ion beam etching machine

      Wet etching equipment

      Ashing equipment

(2) Cleaning system

      Dry etching cleaning equipment

15       UV/O<sub>3</sub> cleaning machine

         Ion beam cleaning machine

         Laser beam cleaning machine

         Plasma cleaning machine

         Gas etching cleaning machine

20       Extractive cleaning equipment

         Soxhlet extractive cleaning machine

         High temperature high pressure extractive cleaning  
machine

         Microwave extractive cleaning machine

25       Supercritical extractive cleaning machine

(3) Exposing system

      Stepper

      Coater and developer

(4) Polishing system

30       CMP equipment

(5) Film forming system

CVD equipment

Sputtering equipment

(6) Diffusion and ion implantation system

Oxidation and diffusion equipment

5 Ion implantation equipment

The present invention is then explained by means of examples and preparation examples, but is not limited to them.

#### PREPARATION EXAMPLE 1

10 (Preparation of emulsion of fluorine-containing elastomer particles)

A 47-liter stainless steel autoclave having no ignition source was charged with 30 liter of pure water, 300 g of  $C_7F_{15}COONH_4$  as an emulsifying agent and 2.7 g of disodium phosphate $\cdot 12H_2O$  as a pH control agent. After the inside of a system was sufficiently replaced with nitrogen gas and deairing was carried out, the autoclave was heated to 50°C with stirring at 200 rpm, and a gas mixture of tetrafluoroethylene (TFE)/perfluoro(methyl vinyl ether) (PMVE) (TFE/PMVE=24/76 in mole ratio) was introduced so that the inside pressure became 1.18 MPa. Then, 100 ml of an aqueous solution of ammonium persulfate (APS) having a concentration of 55.8 mg/ml was introduced with pressurized nitrogen gas to initiate a reaction.

With the advance of the polymerization, at the time when the inside pressure was lowered to 1.08 MPa, 62.3 g of di-iodine compound  $[I(CF_2)_4I]$  was introduced with pressurized nitrogen gas. Then, 60 g of TFE was introduced at its self-pressure and 60 g of PMVE was introduced with a plunger pump so that the inside pressure became 1.18 MPa. Thereafter, with the advance of the reaction, pressurized TFE and PMVE were introduced similarly in the same manner. Thus, increasing and decreasing of the inside pressure were repeated between 1.08 MPa and 1.18 MPa, and at the time when a total amount of TFE and PMVE reached 6.5 kg, 7.8 kg, 9.1 kg and 10.4

kg, respectively, 25.6 g of an iodine compound  $\text{CF}_2=\text{CFOCF}_2\text{CF}_2\text{CH}_2\text{I}$  was introduced with pressurized nitrogen gas. Every 12 hours after starting of the polymerization, 20 ml of an aqueous solution of APS having a concentration of 52.5 mg/ml was introduced with pressurized  
5 nitrogen gas.

When a total amount of the introduced TFE and PMVE reached 13 kg, i.e., 33 hours after starting of the polymerization reaction, the autoclave was cooled and ~~an~~ the un-reacted monomer was released to obtain an emulsion (A-1) of fluorine-containing  
10 elastomer particles (average particle size: 70 nm) having a solid concentration of 27.5 % by weight.

A part of the emulsion was sampled and nitric acid was added thereto for coagulation. The coagulated product was washed and dried to obtain elastomer particles. The Mooney viscosity  $\text{ML}_1 + 10$   
15 (100°C) of the elastomer was 42, and according to  $^{19}\text{F}$ -NMR analysis, the components thereof was TFE/PMVE=62/38 (% by mole). The glass transition temperature  $T_g$  (center value) measured according to DSC was -3°C.

20

## PREPARATION EXAMPLE 2

(Preparation of emulsion of fluorine-containing resin fine particles)

A 6-liter stainless steel autoclave having no ignition source was charged with 3 liter of pure water, 30 g of  $\text{C}_3\text{F}_7\text{OCF}(\text{CF}_3)\text{CF}_2\text{OCF}(\text{CF}_3)\text{COONH}_4$  as an emulsifying agent and 0.27  
25 g of disodium phosphate·12H<sub>2</sub>O as a pH control agent. After the inside of a system was sufficiently replaced with nitrogen gas and deairing was carried out, the autoclave was heated to 80°C with stirring at 600 rpm, and a gas mixture of tetrafluoroethylene (TFE)/perfluoro(methyl vinyl ether) (PMVE) (TFE/PMVE=88/12 in mole ratio) was introduced  
30 so that the inside pressure became 0.20 MPa. Then, 4 ml of an aqueous solution of ammonium persulfate (APS) having a



concentration of 2.5 mg/ml was introduced with pressurized nitrogen gas to initiate a reaction.

With the advance of the polymerization, at the time when the inside pressure was lowered to 0.15 MPa, ~~a.~~ A gas mixture of TFE/PMVE (TFE/PMVE=95/5 in mole ratio) was then introduced with pressurized nitrogen gas so that the inside pressure became 0.20 MPa. Thereafter, with the advance of the reaction, pressurized TFE and PMVE gas mixture (95/5 in mole ratio) was introduced similarly. Thus, increasing and decreasing of the inside pressure were repeated between 0.15 MPa and 0.20 MPa.

When a total amount of the introduced TFE and PMVE reached 331 g, i.e., 4.5 hours after starting of the polymerization reaction, the autoclave was cooled and ~~an~~ the un-reacted monomer was released to obtain an emulsion (B-1) of fluorine-containing resin fine particles (average particle size: 44 nm) having a solid concentration of 9.7 % by weight. The average particle size was ~~measured-determined~~ by mixing 120 mg of the emulsion and 4.4 g of dimethylsulfoxide and ~~measuring-measured~~ with LPA-3000, 3100 available from OTSUKA DENSHI KABUSHIKI KAISHA.

A part of the emulsion was sampled and nitric acid was added thereto for coagulation. The coagulated product was washed and dried to obtain a white fluorine-containing resin fine powder. The melt flow rate MFR of the fluorine-containing resin ~~was un-measurable~~ could not be measured under conditions of maintaining at 372°C for five minutes. According to <sup>19</sup>F-NMR analysis, the components thereof was TFE/PMVE=94.5/5.5 (% by mole). The melting point measured according to DSC was 290°C.

### PREPARATION EXAMPLE 3

(Preparation of emulsion of fluorine-containing resin fine particles)

A 6-liter stainless steel autoclave having no ignition source

was charged with 3 liter of pure water, 30 g of  $\text{C}_3\text{F}_7\text{OCF}(\text{CF}_3)\text{CF}_2\text{OCF}(\text{CF}_3)\text{COONH}_4$  as an emulsifying agent and 0.27 g of disodium phosphate  $\cdot 12\text{H}_2\text{O}$  as a pH control agent. After the inside of a system was sufficiently replaced with nitrogen gas and deairing  
5 was carried out, the autoclave was heated to  $80^\circ\text{C}$  with stirring at 600 rpm, and a gas mixture of tetrafluoroethylene (TFE)/perfluoro(methyl vinyl ether) (PMVE) (TFE/PMVE=88/12 in mole ratio) was introduced so that the inside pressure became 0.20 MPa. Then, 4 ml of an aqueous solution of ammonium persulfate (APS) having a  
10 concentration of 2.5 mg/ml was introduced with pressurized nitrogen gas to initiate a reaction.

With the advance of the polymerization, at the time when the inside pressure was lowered to 0.15 MPa, 2.92 g of a di-iodine compound  $[\text{I}(\text{CF}_2)_4\text{I}]$  was introduced with pressurized nitrogen gas and  
15 then, a gas mixture of TFE/PMVE (TFE/PMVE=95/5 in mole ratio) was introduced with pressurized nitrogen gas so that the inside pressure became 0.20 MPa. Thereafter with the advance of the reaction, pressurized TFE/PMVE gas mixture (95/5 in mole ratio) was introduced similarly. Thus, increasing and decreasing of the inside  
20 pressure were repeated between 0.15 MPa and 0.20 MPa.

When a total amount of the introduced TFE and PMVE reached 327 g, i.e., 9.9 hours after starting of the polymerization reaction, the autoclave was cooled and ~~an~~the un-reacted monomer was released to obtain an emulsion (B-2) of fluorine-containing resin  
25 fine particles (average particle size: 44 nm) having a solid concentration of 9.4 % by weight.

A part of the emulsion was sampled and nitric acid was added thereto for coagulation. The coagulated product was washed and dried to obtain a white fluorine-containing resin fine powder. The  
30 melt flow rate MFR of the fluorine-containing resin was 21.1 g/10 min under conditions of maintaining at  $372^\circ\text{C}$  for five minutes. According

to  $^{19}\text{F}$ -NMR analysis, the components thereof was TFE/PMVE=94.9/5.1 (% by mole). The melting point measured according to DSC was 280.7°C.

#### PREPARATION EXAMPLE 4

(Preparation of emulsion of fluorine-containing elastomer particles)

A 3-liter stainless steel autoclave having no ignition source was charged with 1 liter of pure water, 10 g of  $\text{C}_3\text{F}_7\text{OCF}(\text{CF}_3)\text{CF}_2\text{OCF}(\text{CF}_3)\text{COONH}_4$  as an emulsifying agent and 0.09 g of disodium phosphate·12H<sub>2</sub>O as a pH control agent. After the inside of a system was sufficiently replaced with nitrogen gas and deairing was carried out, the autoclave was heated to 53°C with stirring at 600 rpm, and a gas mixture of tetrafluoroethylene (TFE)/perfluoro(methyl vinyl ether) (PMVE) (TFE/PMVE=25/75 in mole ratio) was introduced so that the inside pressure became 0.78 MPa. Then, 20 ml of an aqueous solution of ammonium persulfate (APS) having a concentration of 264 mg/ml was introduced with pressurized nitrogen gas to initiate a reaction.

With the advance of the polymerization, at the time when the inside pressure was—lowered to 0.69 MPa, 2.2 g of  $\text{CF}_2=\text{CFOCF}_2\text{CF}(\text{CF}_3)\text{OCF}_2\text{CF}_2\text{CN}$  (CNVE) was introduced with pressurized nitrogen gas. Then, 4.7 g of TFE and 5.3 g of PMVE were introduced at the respective self-pressures. Thereafter, with the advance of the reaction, pressurized TFE and PMVE were introduced similarly. Thus, increasing and decreasing of the inside pressure were repeated between 0.69 MPa and 0.78 MPa, and in addition, at the time when a total amount of the introduced TFE and PMVE reached 70 g, 2.2 g of CNVE was introduced with pressurized nitrogen gas.

When a total amount of the introduced TFE and PMVE reached 130 g, i.e., six hours after starting of the polymerization reaction, the autoclave was cooled and ~~an~~the un-reacted monomer

was released to obtain 1,160 g of emulsion (A-2) of fluorine-containing elastomer particles having a solid concentration of 11.3 % by weight.

Then, 100 g of the obtained emulsion was diluted with 300 g of water, and added slowly to 280 g of an aqueous solution of 3.5 % by weight of hydrochloric acid with stirring. After the addition, stirring was carried out for five minutes and a coagulated product was filtrated. The obtained elastomer particles were further washed with 200 g of HCFC-141b and filtrated. After the washing with HCFC-141b and the filtration were repeated four times, vacuum drying was carried out at 60°C for 72 hours to obtain 11.2 g of fluorine-containing elastomer.

As a result of  $^{19}\text{F}$ -NMR analysis, the obtained fluorine-containing elastomer was one comprising TFE/PMVE/CNVE = 60.4/38.9/0.7 (% by mole).

#### EXAMPLE 1

The emulsion of the fluorine-containing elastomer particles (A-1) prepared in Preparation Example 1 and the emulsion of the fluorine-containing resin fine particles (B-1) prepared in Preparation Example 2 were mixed in the amounts of 1,236 g and 619 g, respectively (solid content ratio: fluorine-containing elastomer/fluorine-containing resin = 85/15 in weight ratio), and the mixture was added dropwise over 10 minutes to 411 g of a 9 % aqueous solution of nitric acid being stirred and thus subjected to coagulation. The obtained co-coagulate was washed with water and dried to obtain an elastomer composition comprising the fluorine-containing resin fine particles finely dispersed in the fluorine-containing elastomer.

When the elastomer composition was subjected to DTA measurement, there was ~~recognized an absorption~~ at 272.6°C ~~an absorption considered which is believed~~ to be derived from the

fluorine-containing resin. The composition was transparent (haze value: 12 %).

(Measurement of haze value)

A 0.7 mm thick sample is produced from the elastomer composition at a melting temperature thereof (60° to 150°C) by applying a pressure thereto if necessary. A haze value of the sample is measured with a direct current haze meter (a measuring device according to JIS K7105 and ASTM D 1003 available from TOYO SEIKI KABUSHIKI KAISHA).

A haze value of a crosslinked molded article described hereinbelow is measured by using a 2 mm thick sample sheet produced by crosslinking and molding the elastomer composition.

Fig. 1 shows a photograph of the elastomer composition taken at  $\times 20000$  magnification with a transmission type electron microscope (TEM) by a replica method. In Fig. 1, convex portions in the form of particle are fine particles of the fluorine-containing resin. It can be seen that many fluorine-containing resin fine particles (average particle size: about 40 nm) are finely dispersed uniformly. This finely dispersed state clearly differs from that of a composition in a TEM photograph (Fig. 2) obtained in Comparative Example 3 described hereinbelow.

## EXAMPLE 2

Co-coagulation was carried out in the same manner as in Example 1 except that ~~a~~the mixing amount of the emulsion (B-1) of the fluorine-containing resin fine particles was changed to 412 g, to obtain an elastomer composition (solid content ratio: fluorine-containing elastomer/fluorine-containing resin = 90/10 in weight ratio). The composition was transparent (haze value: 12 %).

## EXAMPLE 3

Co-coagulation was carried out in the same manner as in

Example 1 except that ~~a~~the mixing amount of the emulsion (B-1) of the fluorine-containing resin fine particles was changed to 206 g, to obtain an elastomer composition (solid content ratio: fluorine-containing elastomer/fluorine-containing resin = 95/5 in weight ratio).

5 The composition was transparent (haze value: 11 %).

#### EXAMPLE 4

Co-coagulation was carried out in the same manner as in Example 1 except that 638 g of the emulsion (B-2) of the fluorine-  
10 containing resin fine particles obtained in Preparation Example 3 was mixed instead of the emulsion (B-1) of the fluorine-containing resin fine particles, to obtain an elastomer composition (solid content ratio: fluorine-containing elastomer/fluorine-containing resin = 85/15 in weight ratio). The composition was transparent (haze value: 12 %).

15

#### EXAMPLE 5

To 100 parts by weight of the elastomer composition prepared in Example 1 were mixed 1 part by weight of 2,5-dimethyl-2,5-di(t-butylperoxy)hexane as a crosslinking agent and 1 part by  
20 weight of triallyl isocyanurate as a crosslinking accelerator. The mixture was kneaded with an open roll to obtain a crosslinkable elastomer composition.

Vulcanizability of the crosslinkable elastomer composition was determined by a method described hereinbelow. The results are  
25 shown in Table 4.

Further<sub>1</sub> the crosslinkable elastomer composition was subjected to press-crosslinking at 150°C for 30 minutes and then crosslinking in an oven at 180°C for four hours to obtain a crosslinked article. Physical properties in normal state of the crosslinked article  
30 were measured. Also<sub>1</sub> an O-ring (P-24) was produced under the same crosslinking conditions and a compression set thereof was measured. Further<sub>1</sub> a haze value of the crosslinked elastomer sheet (2 mm thick)

was measured. The results are shown in Table 4.  
(Vulcanizability)

A vulcanization curve of each composition for vulcanization is obtained at a temperature shown in Table 4 by using a JSR Curastometer Model II, and a minimum torque, maximum torque, induction time and optimum vulcanization time are obtained therefrom.

(Physical properties in normal state)

100 % modulus, tensile strength, tensile elongation and hardness (JIS A hardness) in normal state (25°C) are measured according to JIS K6301.

(Compression set)

A compression set after allowing to stand at 200°C for 70 hours is measured according to JIS K6301.

15

#### EXAMPLES 6 to 10

A crosslinkable elastomer composition having components shown in Table 4 was prepared in the same manner as in Example 5 and vulcanizability thereof was determined. Also a crosslinked article was produced under the same crosslinking conditions as in Example 5, and physical properties in normal state, compression set and haze value were determined. The results are shown in Table 4.

A filler mixed in Examples 6 to 8 is a silicon oxide (AEROSIL 300 available from NIPPON AEROSIL KABUSHIKI KAISHA).

25

#### COMPARATIVE EXAMPLES 1 to 2

A crosslinkable elastomer composition (haze value: 10 %) was prepared in the same manner as in Example 5 except that fluorine-containing resin fine powder was not mixed. Vulcanizability of the composition was determined, and also a crosslinked article was produced under the same crosslinking conditions as in Example 5, and physical properties in normal state and compression set were

30

determined. The results are shown in Table 4.

### COMPARATIVE EXAMPLE 3

To the emulsion (A-1) of the fluorine-containing elastomer  
5 particles obtained in Preparation Example 1 was added a nitric acid for  
coagulation thereof, and the coagulate was washed and dried to obtain  
elastomer particles. On the other hand, to the emulsion (B-1) of the  
fluorine-containing resin fine particles obtained in Preparation  
Example 2 was added a nitric acid for coagulation thereof, and the  
10 coagulate was washed and dried to obtain a white fluorine-containing  
resin powder. To 85 parts by weight of the elastomer particles was  
dry-blended 15 parts by weight of the fluorine-containing resin powder  
to obtain a white opaque composition (haze value: 82 %). A TEM  
photograph ( $\times 20000$ ) of the composition is shown in Fig. 2. As it is  
15 clear from Fig. 2, an elastomer portion (flat portions at both sides in  
Fig. 2) is separated clearly from a portion where fluorine-containing  
resin fine particles are agglomerated (a portion having a coarse surface  
at a center in Fig. 2).

A crosslinkable elastomer composition was prepared in the  
20 same manner as in Example 5 except that the above-mentioned  
composition was used, and vulcanizability thereof was determined.  
Also a crosslinked article was produced under the same crosslinking  
conditions as in Example 5, and physical properties in normal state,  
compression set and haze value were determined. The results are  
25 shown in Table 4.



TABLE 4

Elastomer composition	Ex. 5	Ex. 6	Ex. 7	Ex. 8	Ex. 9	Ex. 10	Com. Ex. 1	Com. Ex. 2	Com. Ex. 3
Example 1	100	100				100			
Example 2			100						
Example 3				100					
Example 4					100				
Preparation Example 1 (elastomer)							100	100	85
Preparation Example 2 (fluorine- containing resin powder)									15
Crosslinking agent	1	1	1	1	1	1	1	1	1
Crosslinking accelerator	1	3	3	3	1	0.5	1	3	1
SiO <sub>2</sub>	-	3	3	3	-	-	-	3	-

- continued -

- continued -

Vulcanizability (160°)										
Minimum torque (kg)	0.18	0.40	0.27	0.15	0.10	0.17	0.03	0.10	0.06	
Maximum torque (kg)	4.51	7.70	7.20	6.15	5.02	4.12	3.98	6.00	4.71	
Induction time (min)	0.7	0.5	0.5	0.5	0.6	0.7	0.5	0.6	0.5	
Optimum vulcanization time (min)	1.4	1.4	1.2	1.4	1.5	1.4	1.1	1.1	0.9	
Physical properties in normal state										
100 % modulus (MPa)	4.5	13.8	7.4	6.4	3.7	4.5	2.1	4.9	4.7	
Tensile strength (MPa)	20.8	24.9	22.8	16.3	18.3	19.7	12.3	16.5	8.6	
Elongation (%)	202	155	208	152	197	208	186	180	168	
Hardness (JIS A)	71	80	75	71	71	70	60	68	71	
Compression set (%)	33	27	22	12	34	36	18	13	37	
Haze value (%)	41	-	-	-	43	20	-	-	82	

#### EXAMPLE 11

The fluorine-containing elastomer particles (A-2) prepared in Preparation Example 4 and the fluorine-containing resin fine particles (B-1) prepared in Preparation Example 2 were mixed in the amounts of 300 g and 619 g, respectively in a ratio of fluorine-containing elastomer/fluorine-containing resin = 85/15 (weight ratio), and the mixture was added dropwise over 20 minutes for co-coagulation to 981 g of a 9 % aqueous solution of nitric acid being stirred. The obtained coagulate was washed with water and dried to obtain a transparent elastomer composition (haze value: 18 %) comprising the fluorine-containing resin fine particles finely dispersed in the fluorine-containing elastomer.

#### EXAMPLE 12

A crosslinkable elastomer composition was prepared by kneading 1.45 parts by weight of 2,2-bis-[(3-amino-4-phenylamino)phenyl]hexafluoropropane (synthesized by a method described in Journal of Polymer Science, edited by Polymer Chemistry, Vol. 20, 2381 to 2393 (1982)) as a crosslinking agent based on 100 parts by weight of the elastomer composition prepared in Example 11 by using an open roll. Vulcanizability of the obtained crosslinkable elastomer composition which was determined by the above-mentioned method was as follows.

##### Vulcanizability (170°C)

Minimum torque: 0.45 kg

Maximum torque: 2.90 kg

Induction time: 4.4 min

Optimum vulcanization time: 8.7 min

Further, the crosslinkable elastomer composition was subjected to press-crosslinking at 170°C for 15 minutes and crosslinking in an oven at 204°C for 18 hours and then at 288°C for 18

hours to obtain a crosslinked article. Physical properties in normal state of the obtained crosslinked article was also determined in the same manner as above, and also an O-ring (P-24) was produced under the same conditions as above to determine its compression set. The results are as follows.

Physical properties in normal state

100 % modulus: 2.9 MPa

Tensile strength: 19.4 MPa

Elongation: 252 %

Hardness (JIS A): 74

Compression set (200°C, 70 hours, 25 % compression)

Compression set: 15 %

INDUSTRIAL APPLICABILITY

The present invention can provide a composition capable of ~~giving~~ producing an elastomeric molded article excellent in mechanical strength, abrasion resistance, transparency, etc.

ABSTRACT

A transparent elastomer composition is obtained by co-  
coagulation of an emulsion of fluorine-containing resin fine particles  
5 having an average particle size of from 20 to 150 nm and an emulsion  
of elastomer particles, in which the fluorine-containing resin fine  
particles are finely dispersed uniformly in the elastomer ~~is obtained~~.  
The transparent elastomer composition provides an elastomeric molded  
article having excellent mechanical strength, abrasion resistance and  
10 transparency.